



Metal Distribution Characteristics in a Laboratory Waste Incinerator

Chih-Ping Chang^{1,5}, Yun-Hwei Shen¹, I-Cheng Chou², Ya-Fen Wang³, Yi-Ming Kuo^{2*},
Juu-En Chang⁴

¹ Department of Resources Engineering, National Cheng Kung University, 1 University Rd., Tainan City 701, Taiwan

² Department of Safety Health and Environmental Engineering, Chung Hwa University of Medical Technology, 89 Wenhua 1st St., Rende Dist., Tainan City 717, Taiwan

³ Department of Bioenvironmental Engineering, Chung Yuan Christian University, 200 Chung Pei Rd., Chung Li 320, Taiwan

⁴ Department of Environmental Engineering, National Cheng Kung University, 1 University Rd., Tainan City 701, Taiwan

⁵ Environmental Resource Management Research Center, Cheng Kung University, 500 Anming Rd. Sec. 3, Annan Dist., Tainan City 709, Taiwan

ABSTRACT

The metal distribution in the emissions of a laboratory waste incinerator (LWI) was investigated. The input materials and output materials (fly ash, bottom ash, and flue gas) were analyzed to examine the transportation and transmission behavior of heavy metals.

Toxic (As, Cd, Cr, Hg, Ni, and Pb), anthropogenic (Ag, Ba, Cu, Mn, Se, and Zn) and crust (Na) elements were adopted to characterize the distribution properties of the LWI under investigation. The results indicated that Ba, Cr and Hg (80.0, 13.0 and 12.9 mg/L, respectively) were the major metals in the organic liquid waste input, and Na, and Zn (7497 and 349 mg/kg, respectively) were the major ones for medical solid waste. The Na content in the fly ash could be as high as 911574 mg/kg, and this was found to be the major Na emission source according to the emission factors. The mass of Na may be supplied using a NaOH solution injection to neutralize the acidic combustion gas, and it was found that high contents of Na in the flue gas were from the sprinkled NaOH solution. For the output materials, NaCl was found to be the major crystalline form of Na in the ash, based on the results of X-ray diffraction (XRD) patterns. The crust (Na) element was the major element in the bottom and fly ash. The toxic (As, Cr, Ni, and Pb) and anthropogenic (Se) elements were mainly distributed in the second cooling tower ash and baghouse ash in a range of 64.5%–91.9%. The average concentrations of Pb, Cd, and Hg concentrations in the flue gas were all under Taiwan's EPA regulation standards.

Toxicity characteristic leaching procedure (TCLP) data showed that the leachability of As, Cr, Cu, Hg, and Se exceeded the EPA's regulations. It is thus suggested that the ash should be treated by further elutriation or vitrification processes to reduce the TCLP levels in the final products.

Keywords: Incineration; Metal distribution; Laboratory waste; Solid waste.

INTRODUCTION

Because of Waste Disposal Act restrictions in Taiwan, university laboratory waste cannot be treated in municipal solid waste incinerators or using simple laboratory-scale treatment processes. The Sustainable Environment Research Center (SERC) at National Cheng Kung University (NCKU) was thus established to treat laboratory waste with a physical-chemical treatment system, incineration system, and plasma melting vitrification system. The SERC divides

the laboratory waste into 10 categories: organic liquid waste (halogen), organic liquid waste (non-halogen), organic liquid waste (waste oil), cyanide liquid waste, mercury liquid waste, acid liquid waste, alkaline liquid waste, heavy metal liquid waste, medical solid waste, and special waste. Organic liquid waste, partial medical solid waste, and partial special waste are treated in an incinerator. The sludge and ash from the physical-chemical treatment and incineration systems can be further treated by the plasma melting vitrification system to reduce the amount of hazardous materials that have not decomposed completely (Kuo *et al.*, 2010).

Previous studies have shown that municipal solid waste incinerators are the main emissions source of persistent organic pollutants (e.g., polycyclic aromatic hydrocarbons, polychlorinated dibenzo-*p*-dioxins and furans, polychlorinated biphenyls, polybrominated diphenyls ethers), and that ash

* Corresponding author. Tel: +886-6-267 4567 ext. 854;
Fax: +886-6-267 5049
E-mail address: yiming@mail.hwai.edu.tw

should be retreated if it does not meet the regulated limits (Mi *et al.*, 2001; Wang *et al.*, 2005; Lin *et al.*, 2010; Wang *et al.*, 2010; Artha *et al.*, 2011; Chen *et al.*, 2011; Chiu *et al.*, 2011; Tu *et al.*, 2011; Lin *et al.*, 2012; Wang *et al.*, 2012). In addition to organic pollutants, high metal concentrations have been found in MSW incinerator fly ash and bottom ash (Valavanidis *et al.*, 2008; Gidarakos *et al.*, 2009; Zhao *et al.*, 2010). Metal exhibits good leachability under some environmental conditions (Valavanidis *et al.*, 2008), and the metal leachability of laboratory waste incinerator (LWI) ash raises similar environmental concerns to those for ash from an MSW incinerator.

Plastic solid waste (PSW), organic liquid waste (OLW), and medical solid waste (MSW) are the major kinds of laboratory waste that are treated by the incineration system in the SERC laboratory waste treatment plant. In the present study, the targeted laboratory waste incineration process (for PSW, OLW, and MSW) was surveyed for the metal distributions. The metal distributions in the input and output materials of a LWI and their metal leachabilities were examined in this study to clarify the metal behavior during the laboratory waste incineration process.

METHODS

Selected PSW, OLW, and MSW trials were sampled to survey the metal distribution in the SERC LWI. The PSW was the discarded plastic instruments and broken containers from inorganic and organic liquid waste. Dry liquid waste, which may coat PSW, contributes metal and organic compounds during the incineration process. Dry OLW and other chemicals may coat PSW during experiments or waste storage. In this study, OLW included waste oil and organic liquid solvents. PSW and OLW were collected from all the universities that have contracts with the SERC. MSW was collected from the university medical college.

Fig. 1 shows the material flow of the LWI, which is a two-chamber fixed-hearth incinerator with a maximum capacity of 375 kg/hr. It has a solid waste feed inlet and a liquid waste injection nozzle. The temperature of the primary combustion chambers is in a range of 600–800°C. The bottom ash is mainly generated in the primary combustion chamber, and the secondary combustion chamber is designed to heat the vaporized waste to 1050°C for decomposition of organic compounds. The heat value of the injected liquid waste is designed to be in the range of 1500–5500 kcal. The temperatures of the first and second cooling towers are 250°C and 180°C, respectively. The wet scrubber is designed to cool the gas from 180°C to 80°C before it is expelled from the stack. NaOH solution is injected before the cooling towers and wet scrubber to neutralize the pH value of the emission gas and ash. Active carbon is injected into the incinerator (between the second cooling tower and baghouse filter) to adsorb pollutants that are emitted from the combustion chambers. The baghouse filter is designed to trap pollutant-adsorbed active carbon and fly ash from the combustion chamber.

The input materials (i.e., PSW, OLW, and MSW) and output materials (i.e., bottom ash and fly ash) were collected

from the LWI following Taiwan National Institute of Environmental Analysis (NIEA) method R118.02B. The solid specimens were collected with a stainless steel shovel, and stored at 4°C in a sampling bag. The stack flue gas specimens were collected following NIEA method A302.73C (equivalent to U.S. EPA Method 29). The details of this process can be found in our previous report (Kuo *et al.*, 2010).

All the dehydrated solid specimens were pretreated with microwave-assisted acid digestion, following NIEA method R317.10C (equivalent to U.S. EPA Method 3015A). The specimens were digested using a 400-W microwave MARS/MARS Xpress CEM microwave, with an 800 psi limit at 200°C for 15 minutes. The toxicity characteristic leaching procedure (TCLP) was performed to examine the leachability of LWI ash following NIEA method R201.14C (equivalent to U.S. EPA SW846 Method 1311). The absorption solutions of the stack flue gas, digested solutions of solid specimens, and TCLP solutions were totally digested and passed through 0.45- μ m cellulose acetate filters to remove non-metal materials. The specimen solutions were analyzed following U.S. EPA Method 200.7 with inductively coupled plasma-optical emission spectrometry (ICP-OES, VISTA-MPX, Varian) for Ag, As, Ba, Cd, Cr, Cu, Hg, Mn, Na, Ni, Pb, Se, and Zn.

X-ray diffraction (XRD) was conducted using a power diffractometer (Rigaku, Rint 2000) to determine the crystalline phases of LWI ash. A 10% mass ratio of silicon (99+%) was homogeneously mixed with the specimens as an internal standard for crystalline semi-quantitative analysis (Kuo *et al.*, 2008). The XRD intensity was measured in the 2 θ range of 10° to 60° with Ni-filtered Cu K α radiation at 4°/min (Chou *et al.*, 2011). The crystalline phases of the specimens were identified by comparing the peaks and profiles of the XRD data with the powder diffraction file (PDF) from the International Centre for Diffraction Data. A high-resolution thermal field-emission scanning electron microscope (FE-SEM, JSM-7001 type, JEOL) was used to observe the microstructures of the laboratory waste ash.

RESULTS AND DISCUSSION

Metal Content in Input Materials

This study classified metal elements into toxic (As, Cd, Cr, Hg, Ni, and Pb), anthropogenic (Ag, Ba, Cu, Mn, Se, and Zn) and crust (Na) groups (Wang *et al.*, 2010). The metal compositions of the input materials (i.e., PSW, OLW, and MSW) are shown in Fig. 2. The OLW data was analyzed with XRF, and the PSW and MSW data were analyzed with ICP. Zn and Mn are the major metal components in PSW, with concentrations of 47907 and 2675 mg/kg, respectively. The main metal components in OLW are Ba, Cr, and Hg, with concentrations of 80, 13.0, and 12.9 mg/L, respectively, and those in MSW are Na and Zn, with concentrations of 7497 and 349 mg/kg, respectively. Fig. 2 indicates that the metal emission characteristics are dependent on the input material.

Metal Content of Ash

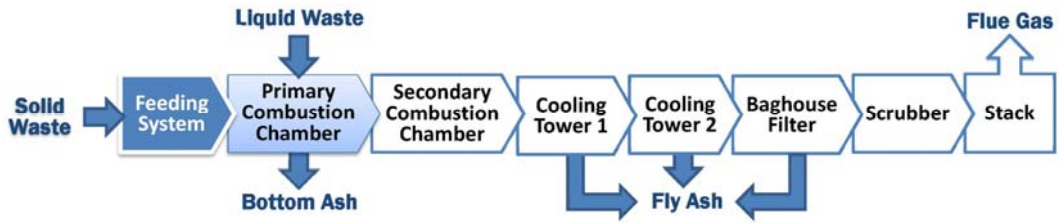


Fig. 1. Material flow of laboratory waste incinerator.

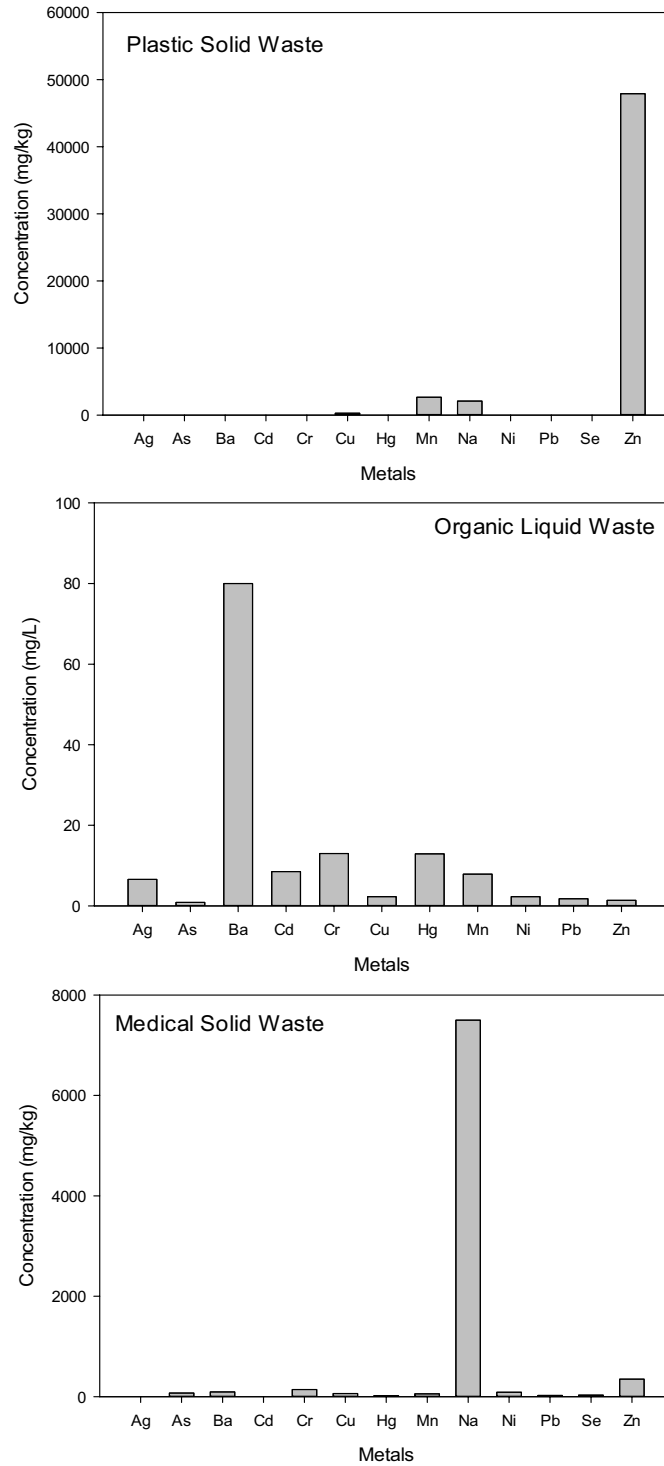


Fig. 2. Metal content of input materials.

The metal content in the PSW, OLW, and MSW ash is shown in Table 1. For the bottom ash from PSW, Na, Ni and Cu are the major metals, with concentrations of 30394, 2762, and 2404 mg/kg, respectively. For the bottom ash from MSW, Na, Cu, and Ba are the major ones, with concentrations of 29117, 5572, and 3742 mg/kg, respectively. No bottom ash was found from the OLW due to the liquid injection.

Na is the major metal in the cooling tower ash. In addition to the input materials, Na may come from the sprinkled NaOH solution, which was adopted to neutralize the acidic combustion gas during the incineration. The metal concentrations in the first and second cooling tower ash show a similar pattern for PSW, OLW, and MSW. Na is the major metal in all cooling tower ashes with 304938 and 278433 mg/kg, 91423 and 911574 mg/kg, and 221662 and 199830 mg/kg for PSW, OLW, and MSW, respectively. In addition to, Na, Hg, and Cr were the two main minor metals for PSW with the concentrations of 1283–2155 mg/kg and 289–203 mg/kg, respectively. Cr and Ni were the two main minor metals for OLW, with the concentrations of 3801–2058 mg/kg and 1594–680 mg/kg, respectively. Finally, Cr and Ni were the two main minor metals in the cooling tower ash for MSW, with concentrations of 384–2812 and 173–4073 mg/kg, respectively.

Na is the major metal in the baghouse ash for PSW, OLW, and MSW, with the concentrations of 208483, 250896, and 73152 mg/kg, respectively. Hg (8618 mg/kg) and Cu (1096 mg/kg) were the minor metals in baghouse ash for PSW. Zn was the minor metal for OLW, and Cr and Ni were that for MSW. The metal composition of the baghouse filter ash is similar to that of the cooling tower ash. Cr was the minor metal in the cooling tower and baghouse ash for PSW and OLW, respectively, and Cr and Ni were the minor metals in the MSW.

Metal Content in Stack Flue Gas

The gaseous and particulate phase metal compositions of stack emissions are shown in Fig. 3. Na is the major emission metal of PSW, OLW, and MSW in the stack flue gas. Hg is the minor one of MSW and OLW, and Zn is that of MSW.

For the gas phase of flue gas, Na is the major metal for PSW and MSW, with concentrations of 898 and 1946 µg/Nm³, respectively. Hg is the major metal for OLW, and the minor metal for PSW and MSW.

In the particulate phase, Na is the dominant metal for PSW, OLW, and MSW, with concentrations of 1047, 376, and 947 µg/Nm³, respectively. The minor metal in the particulate phase for OLW was Hg, and Zn was the minor metal for OLW and MSW, with concentrations of 22.8 and 80.7 µg/Nm³, respectively. According to the waste incinerator air pollutant emissions standards, the regulations for Pb, Cd, and Hg (including their compounds) are 0.5, 0.04, and 0.1 mg/Nm³, respectively. The Pb, Cd, and Hg emission concentrations for the three trials were 8.81–29.7, 0–2.03, and 0.897–293 µg/Nm³, respectively. The metal emission concentrations in the LWI were thus much lower than the standards.

Table 1. Metal content of the bottom ash and fly ash (mg/kg).

Metal	Bottom Ash			1 st Cooling Tower Ash			2 nd Cooling Tower Ash			Baghouse Filter Ash		
	Plastic Solid Waste	Medical Solid Waste	Medical Liquid Waste	Plastic Solid Waste	Organic Liquid Waste	Medical Solid Waste	Plastic Solid Waste	Organic Liquid Waste	Medical Solid Waste	Plastic Solid Waste	Organic Liquid Waste	Medical Solid Waste
Ag	120	21.0	27.3	ND	ND	12.2	25.3	ND	10.0	12.33	ND	19.0
As	85.4	78.8	151	81.8	81.8	76.5	84.4	59.6	54.1	968	85.4	73.9
Ba	677	3,742	40.9	31.2	18.0	18.0	9.63	ND	4.98	30.5	12.7	10.3
Cd	14.0	3.49	73.8	ND	4.77	4.77	12.4	ND	3.55	37.9	ND	11.2
Cr	2,293	1,812	289	3,801	384	384	203	2,058	2,812	266	986	9,550
Cu	2,404	5,572	265	402	46.4	46.4	84.1	165	136	1,906	968	554
Hg	1,113	7.28	1,283	ND	8.06	8.06	2,155	ND	5.16	8,618	ND	8.92
Mn	N.A.	453	N.A.	543	90.3	90.3	N.A.	78.9	405	N.A.	156	1,454
Na	30,394	29,117	304,938	91,423	221,662	221,662	278,433	911,574	199,830	208,483	250,896	73,152
Ni	2,762	2,806	262	1,594	173	173	182	680	4,073	277	458	9,235
Pb	553	70.6	171	54.5	14.4	14.4	171	28.9	14.1	766	364	39.9
Se	373	275	83.5	540	58.7	58.7	54.8	255	422	42.1	150	1,419
Zn	N.A.	1,051	N.A.	652	136	136	N.A.	364	187	N.A.	2,473	571

N.A.: not available; ND: not detectable.

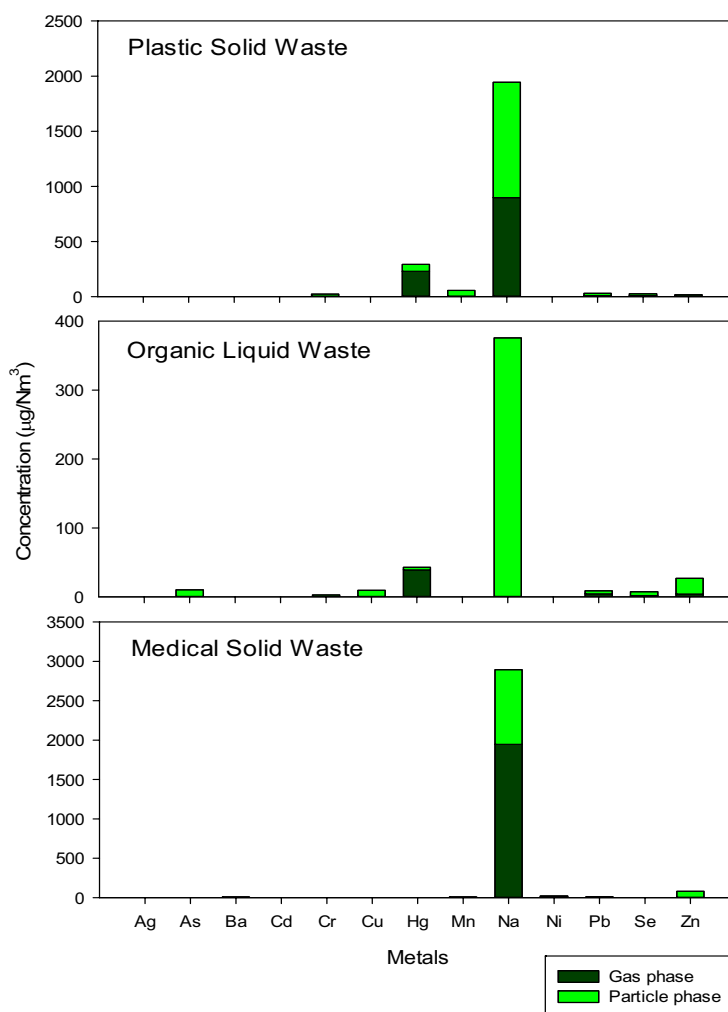


Fig. 3. Gas/particle phase distribution of metals in the flue gas stack.

As, Cd, Hg, and Se are the possible vaporized metals within the range of the incineration temperature (600–800°C). Comparing the melting and boiling points of the major emission metals (Hg, Na, and Zn), the high contents of Na in the flue gas should be from the sprinkled NaOH solution, because the boiling points of Na (886°C) and Zn (907°C) are between 800°C (first combustion chamber) and 1050°C (second combustion chamber). However, the content of Zn in the flue gas was much lower than that of Na, which means the high Na content was from other source rather than the input material.

Incinerator Emission Factors

The metal emission factors (EFs) for incinerator emission sources are shown in Fig. 4. Bottom ash contributes the most metal species among these metal emission sources. The average values and relative standard deviations (RSDs) of the major EFs of Mn and Zn from bottom ash were 68689 (141%), and 132456 (141%) mg/kg-input, respectively. Na was found to be the dominant emission metal in the first and second cooling tower ash, baghouse filter ash, and flue gas. The EFs for Na from the first cooling tower ash, second cooling tower ash, baghouse ash, and stack flue gas

were 1638 mg/kg-input (62.1%), 9946 mg/kg-input (156%), 2936 mg/kg-input (140%), and 148 mg/kg-input (124%), respectively.

The total EFs of the metals were calculated by summing the EFs from each metal emission source. The average EFs of Zn, Mn, and Na in the LWI were 89143, 45867, and 17075 mg/kg-input, respectively. Compared with the toxic, anthropogenic, and crust metals in various output media, anthropogenic and crust metals are the primary and secondary contributors to the bottom ash, with averages of 62.0% and 32.6%. Crust metal is the dominant contributor to the first and second cooling towers, baghouse, and flue gas, with the averages of 95.9%, 97.8%, 87.0%, and 86.3%, respectively.

Metal Emission Distribution in the Output of the Incinerator

The output metal mass emission distributions from the first cooling tower, second cooling tower, baghouse filter, bottom ash, and stack flue gas for PSW, OLW, and MSW are shown in Fig. 5. For PSW, bottom ash was found to be the major contributor medium to the total metal emissions of all the analysed metals in the range of 69.0%–99.9%.

In the case of OLW, the first cooling tower ash was the

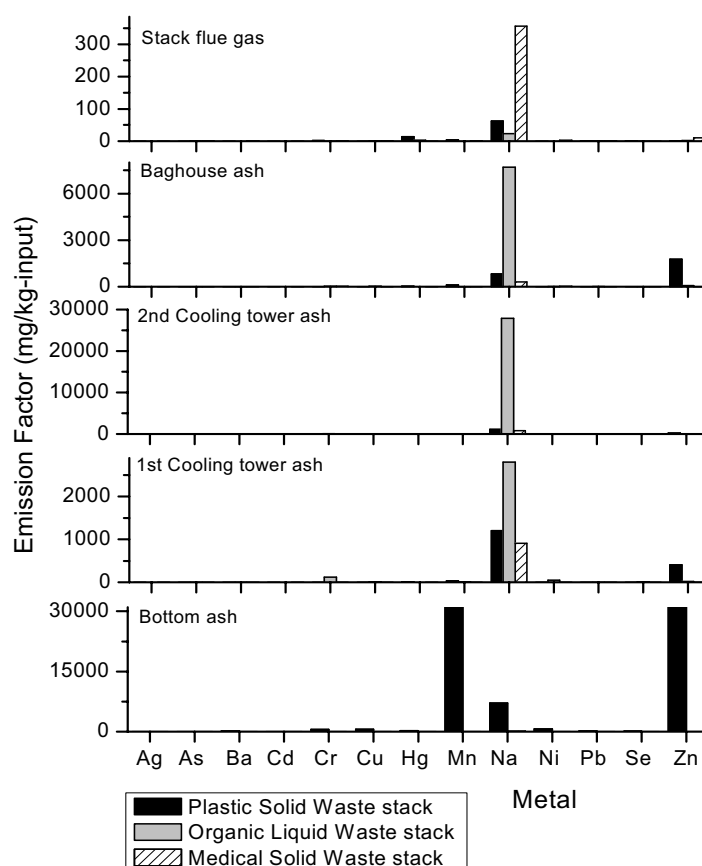


Fig. 4. Metal emission factors of incinerator emission sources.

main emission source of Ba, Cr, Mn, Ni, and Se, in the range of 55.5%–71.1%. Baghouse filter ash was the major emission source of Cu, Pb, and Zn, accounting for 62.3%, 78.2%, and 69.8%, respectively. Hg was mainly emitted from the stack flue gas, As was mainly emitted from the first cooling tower and baghouse filter ash, and Na was mainly emitted from the second cooling tower ash.

In the case of MSW, bottom ash was found to be the major emission source of Ag, Ba, and Cu, in the range of 86.2%–94.8%. Baghouse filter ash was the major source of Cr, Mn, Ni, and Se, in the range of 54.4%–64.9%, and stack flue gas was the major source of As, Cd, Hg, Pb, and Zn in the range of 36.4%–66.4%. The first and second cooling towers were the major sources of Na (69.0%).

Generally speaking, bottom ash was the major emission source for most metals in the PSW trials. This means that most metals remained in the solid or liquid phases, rather than being vaporized into a gas in the combustion chamber. The first and second cooling tower ash was the major source of most metals in the OLW trial. In the case of OLW and MSW, most of the Hg was emitted from the stack flue gas. For MSW trial, bottom ash was the dominant medium for Ag, Ba, and Cu, while the flue gas is the dominant medium for As, Cd, Hg, Pb, and Zn. This shows that the distribution of metals in various media had a correlation with the metals' boiling points, except for Pb. Because Pb is not a potential metal in the gas phase, the Pb found may be from the interference of the memory effect.

TCLP of Output Ash

The toxicity characteristic leaching procedure (TCLP) results for the three trials are shown in Table 2. According to the EPA's regulations, the standards for Ag, As, Ba, Cd, Cr, Cu, Hg, and Se are 5.0, 5.0, 100, 1.0, 5.0, 15.0, 0.2, and 1.0 mg/L, respectively. It can be seen that the TCLP of PSW bottom ash shows a high Hg concentration of 0.218, which is 1.09 times the regulation limit, and that of MSW shows high Cu, Hg, and Se concentrations, which are 1.76, 1.55, and 5.86 times the regulation limits, respectively. For the first cooling tower, the Cr, Hg, and Se concentrations of OLW exceeded the standards, with 12.5, 4.38, and 5.89 times the regulation limits, respectively. For the second cooling tower, the Hg concentration for PSW was 1.09 times the regulation limit. For the baghouse filter ash, Hg exceeded the standard in all trials, with 6.05 (PSW), 2.815 (OLW), and 20.4 (MSW) times the regulation limits, respectively. As was 5.7 times the regulation limit for the PSW baghouse filter, while Cr, Cu, and Se, which were 37.0, 2.39, and 141 times the regulation limits, respectively. Generally speaking, Hg had the highest leachability, followed by Se, Cr, Cu, and As.

Microstructure of Fly Ash and Bottom Ash

The microstructures of fly ash and bottom ash are shown in Fig. 6 (SEM) and Fig. 7 (XRD). The 5000x magnification the SEM micrographs shows that cubic crystal was the major surface structure for the cooling tower and baghouse

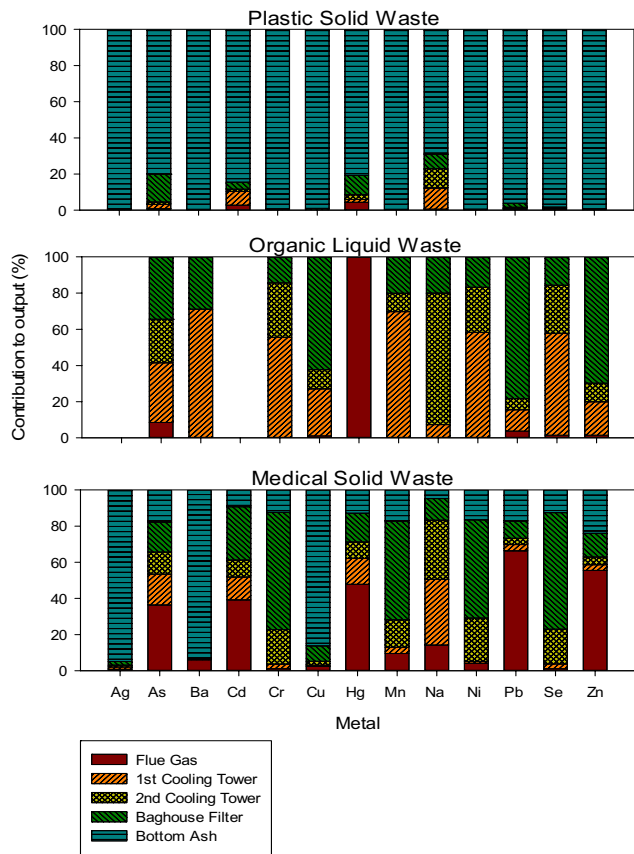


Fig. 5. Mass distribution of metals in output ash and flue gas.

ashes, while the surface structure of bottom ash is an amorphous-like structure without significant crystallinity.

The crystalline phase of the salt was obtained from the XRD analysis. The pure Si (added for semi-quantity) is labelled with a reverse triangle (∇) in Fig. 7. For the cooling tower and baghouse ashes, NaCl (*) is the major salt, which is consistent with the high concentration of Na from the ICP-OES data. It can be seen that the SiO₂ is the major crystalline phase in the bottom ash, which indicates that glassy waste was the primary source of bottom ash.

NaCl has been found to be an accelerator at low levels but a retarder at high levels with regard to the strength development of hydration in ordinary Portland cement (Brough et al., 2000). In addition, the structural integrity of stabilization/solidification samples has been shown to be affected by the leaching of soluble salts from air pollution control residues (Lampris et al., 2009). Hence, the process of NaCl removal (e.g., elutriation) should be taken into account before stabilization/solidification or other further advanced treatment.

CONCLUSIONS

The LWI EFs show a high deviation because of the uncertain composition of the input materials. The crust element (Na) is the major element in all ash. The EFs of individual metals for various output media indicate that the metal emission characteristics of the first and second

Table 2. Toxicity characteristic leaching procedure (TCLP) metal content in the output ash (mg/L).

Metal	TCLP Standard	Bottom Ash			1 st Cooling Tower Ash			2 nd Cooling Tower Ash			Baghouse Filter Ash		
		Plastic Solid Waste	Medical Solid Waste	Organic Liquid Waste	Plastic Solid Waste	Medical Solid Waste	Organic Liquid Waste	Plastic Solid Waste	Medical Solid Waste	Organic Liquid Waste	Plastic Solid Waste	Medical Solid Waste	Organic Liquid Waste
Ag	5.0	ND	0.013	0.287	0.695	0.147	0.325	0.147	0.147	ND	0.147	ND	0.585
As	5.0	0.0806	0.156	0.653	ND	ND	0.0757	ND	ND	0.0662	ND	0.0662	ND
Ba	100	0.129	0.042	ND	0.117	0.003	ND	ND	0.003	ND	ND	ND	ND
Cd	1.0	ND	0.007	0.210	0.141	0.057	ND	ND	0.057	0.0430	0.0796	0.0430	0.405
Cr	5.0	0.0327	0.070	0.0421	0.115	1.74	0.248	0.248	1.74	0.0133	ND	0.0133	185
Cu	15.0	0.0134	26.4	3.13	0.099	1.33	1.61	1.61	1.33	0.0134	2.064	6.72	35.9
Hg	0.2	0.218	0.310	0.0253	0.054	1.73	0.173	0.173	1.73	0.218	1.21	0.563	4.08
Mn	--	0.123	0.440	11.1	2.16	14.7	ND	ND	14.7	ND	0.422	3.09	107
Na	--	20,644	462	26,510	158,379	18,590	35,859	35,859	18,590	31,605	31,605	23,237	1,574
Ni	--	0.453	3.03	4.08	1.96	224	10.7	10.7	224	2.22	2.22	13.6	824
Pb	--	0.0131	0.008	0.140	0.030	0.020	ND	ND	0.020	0.0301	0.0301	0.0220	0.312
Se	1.0	0.103	5.86	0.199	0.018	0.749	0.136	0.136	0.749	0.286	0.286	0.0824	141
Zn	--	ND	5.95	5.20	3.32	5.36	5.43	5.43	5.36	0.164	0.164	27.1	36.9

N.A.: not available; ND: not detectable.

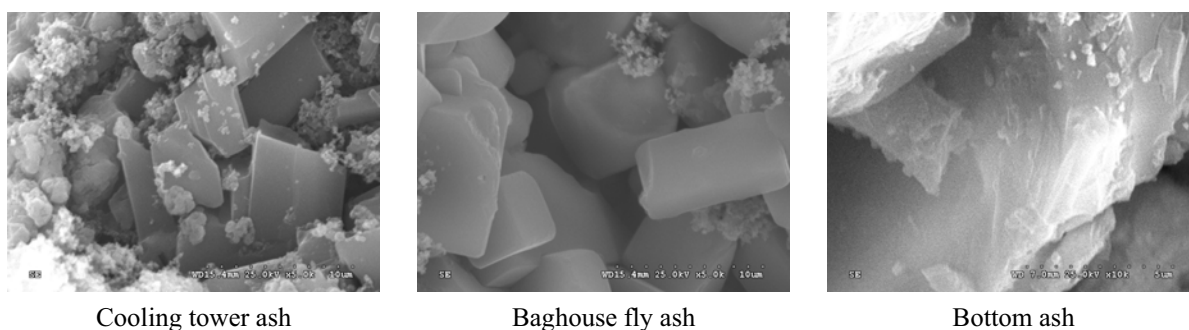


Fig. 6. SEM micrographs of laboratory waste incinerator ash.

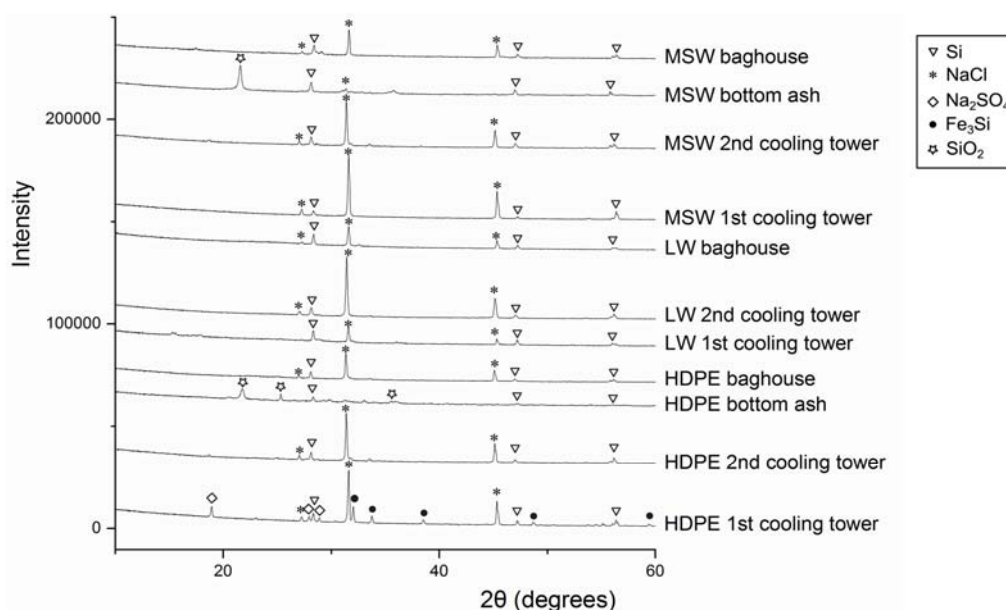


Fig. 7. XRD patterns of laboratory waste incinerator ash.

cooling towers are similar to those of the baghouse filter. Bottom ash was the main metal emission source for PSW. The flue gas concentrations of Pb, Cd, and Hg were much lower than the waste incinerator heavy metal air pollutant emissions standards, except the gaseous Hg in the PSW trial. For MSW, the distribution of metals in various media had a correlation with the metal boiling point

The leachabilities of As, Cr, Cu, Hg, and Se exceeded the TCLP regulation limits. NaCl was identified as the main material that coated LWI ash. The ash from the LWI should be further treated (e.g., vitrification or elutriation processes) for metal and salt separation before final treatment.

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